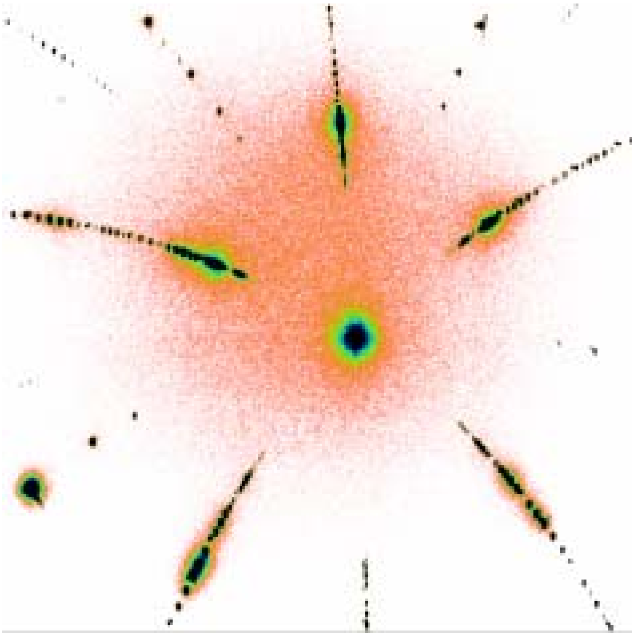
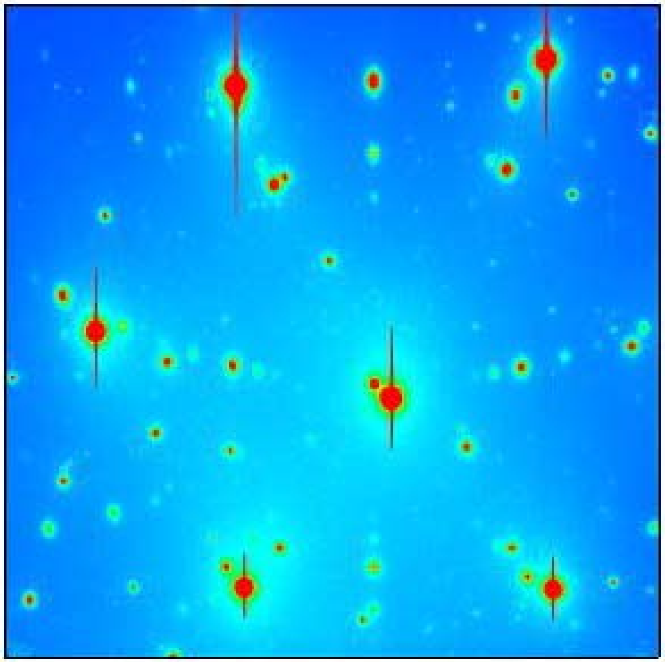
Three-dimensional polychromatic microdiffraction studies of mesoscale structure and dynamics

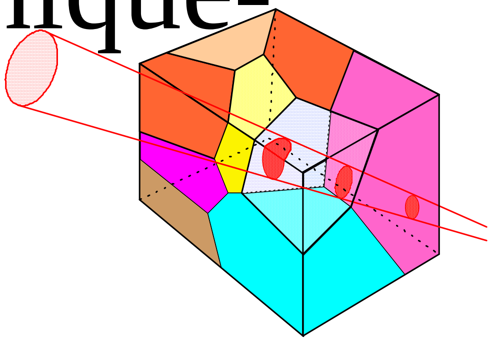
Gene Ice

Oak Ridge National Laboratory





3D polychromatic microdiffraction important emerging technique-

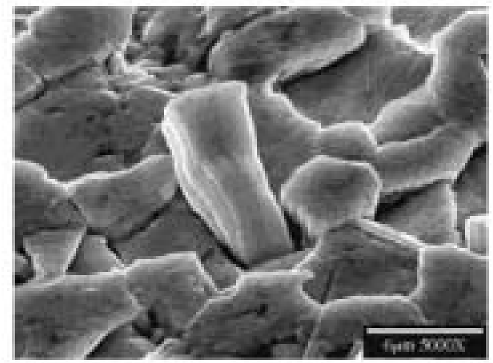


- Fundamentally new direction in materials research
- Unprecedented-direct test of mesoscale modeling
- Addresses long-standing fundamental problems

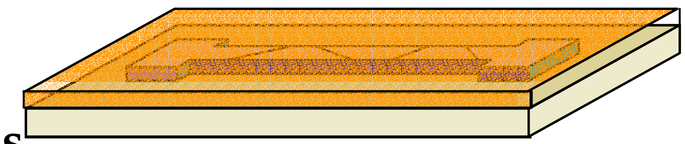
- Programmatic effort only possible at APS
  - requires synchrotron radiation - advanced x-ray optics
  - emerging capabilities only possible with intense 3rd generation source and continued progress

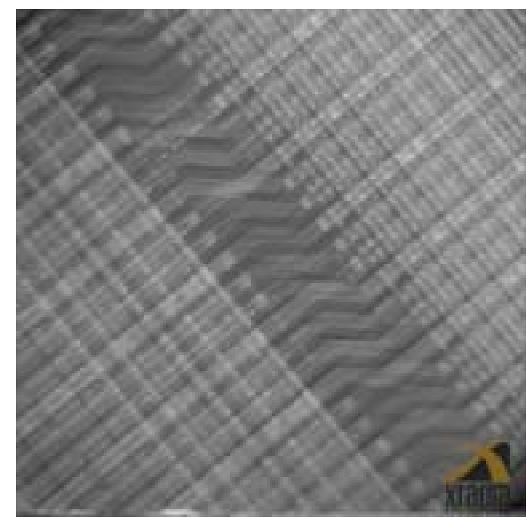
Will become an essential technique for x-ray synchrotron sources.

Virtually all materials influenced by structure and properties at *mesoscale*

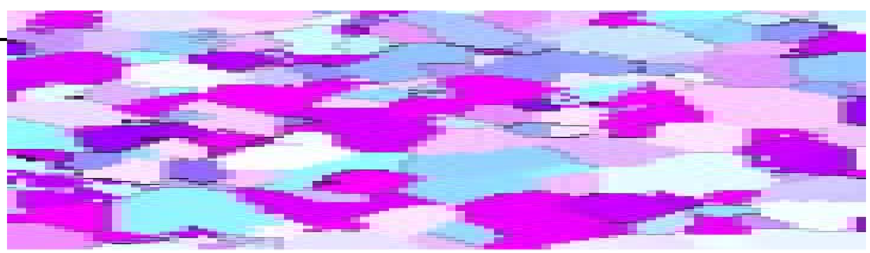


- **Electronic/electro-optic materials**
  - **Thin polycrystalline films/implanted layers defects/anisotropic domains**





- **Structural materials**

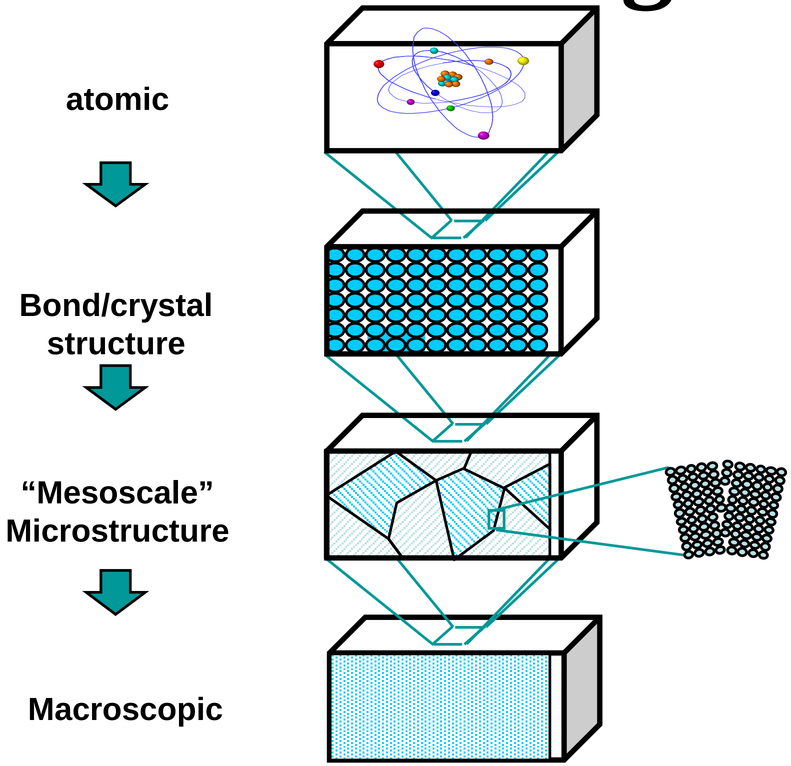


Yet most information -surfaces or property averages

Importance of mesoscale structure/ dynamics→multiscale modeling

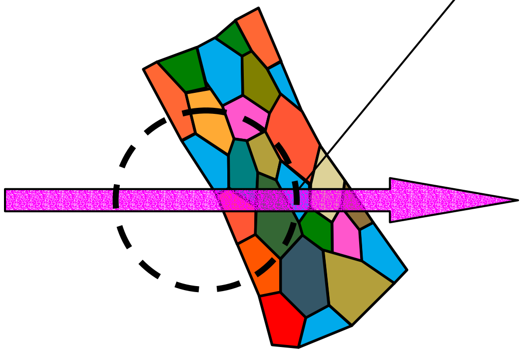
- 0.1-100 μm
  - Too *large* - molecular dynamics
  - Too *small* - average behavior

- Models need guidance
  - Grain boundary structure/properties
  - 3D Deformation
  - Elastic response of grain-boundary networks



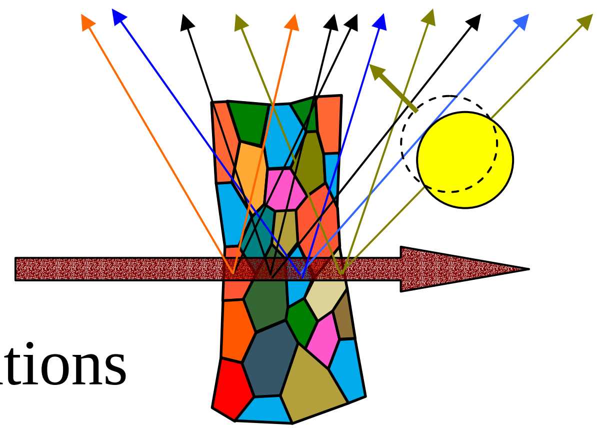
Experimental measurements are essential to guide/test mesoscale modeling

Polychromatic microdiffraction unique advantages



- ***No*** sample rotation-   high spatial resolution

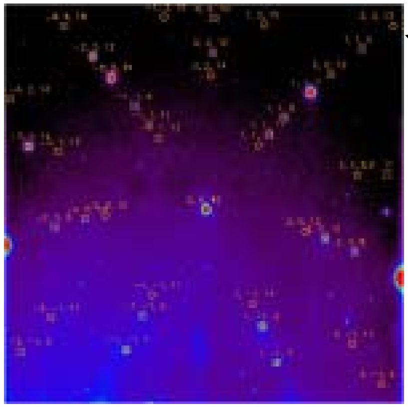
- ***Single crystal*** information from every subgrain volume.
J. S. Chung and G.E. Ice J. Appl. Phys. 86 (1999).



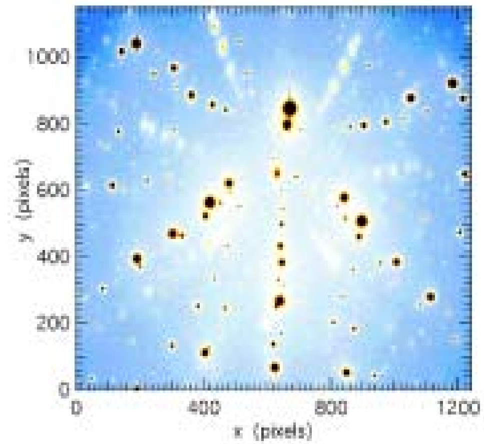
- Differential aperture microscopy ***decodes Laue patterns*** along beam
B. Larson et al. Nature 415 (2002).

Allows investigation of mesoscale correlations

3DPMD correlates mesoscale structural heterogeneities and driving forces

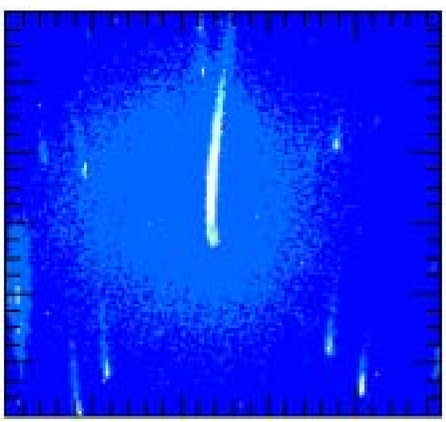


Phase/phase boundaries

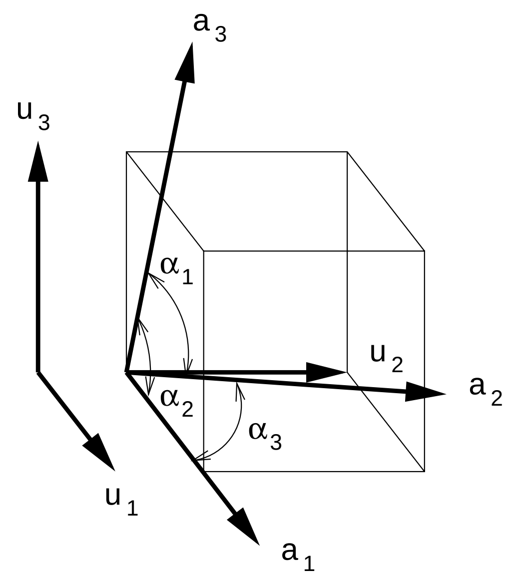


Texture (0.01º)/grain boundaries

Elastic strain (1×10^{-4})



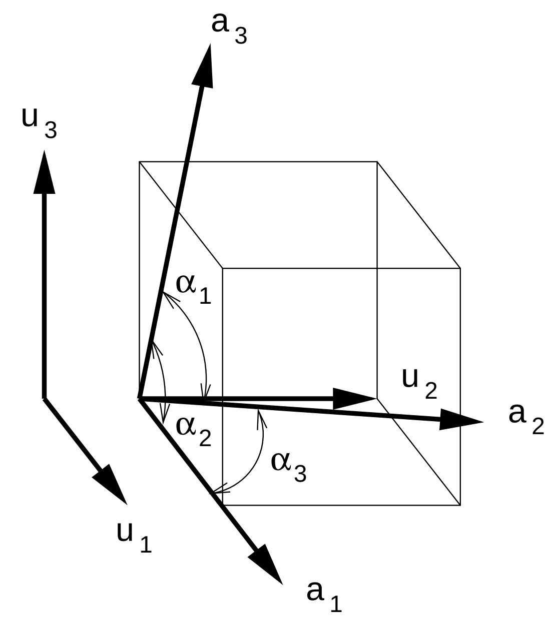
Deformation



Strain is derived from unit cell parameters

$$A_{Meas} = TA_0$$

$$_{ij} = (T_{ij}+T_{ji})/2 - I_{ij}.$$



Accurate measurements require absolute calibration

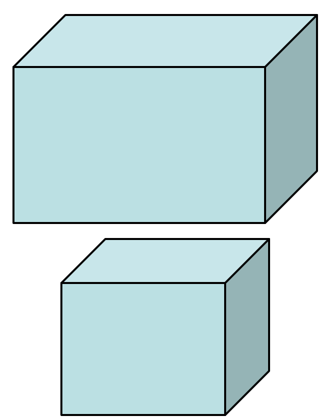
monochromator energy to ~1 eV

CCD to 0.2 pixels ~0.01 degrees

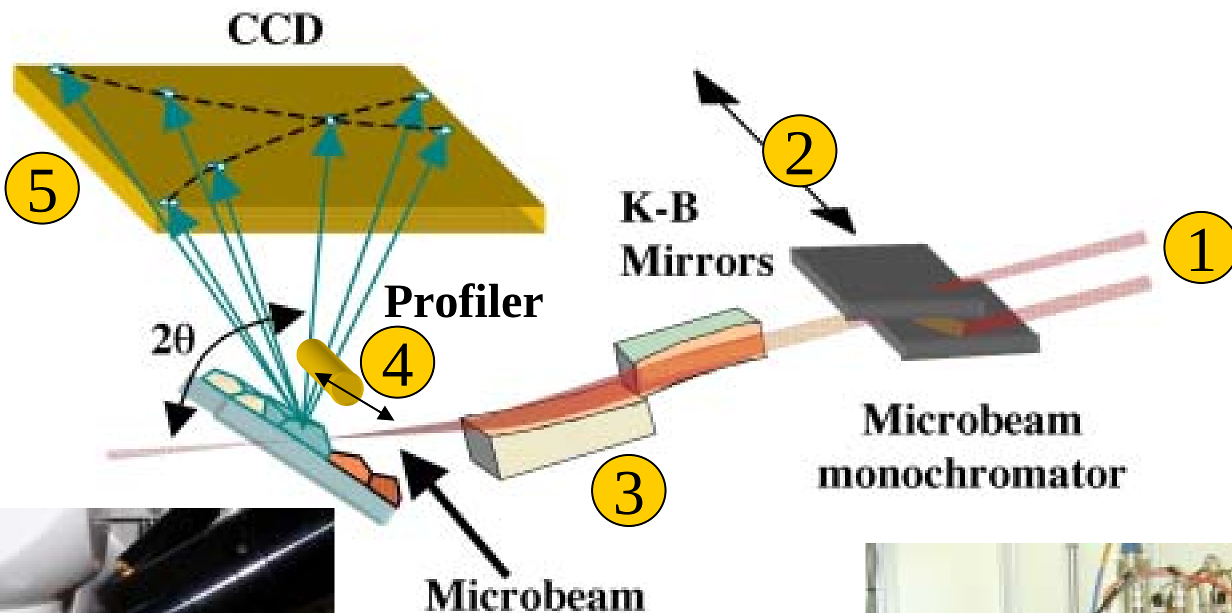
Deviatorial strain tensor from single crystal Laue pattern

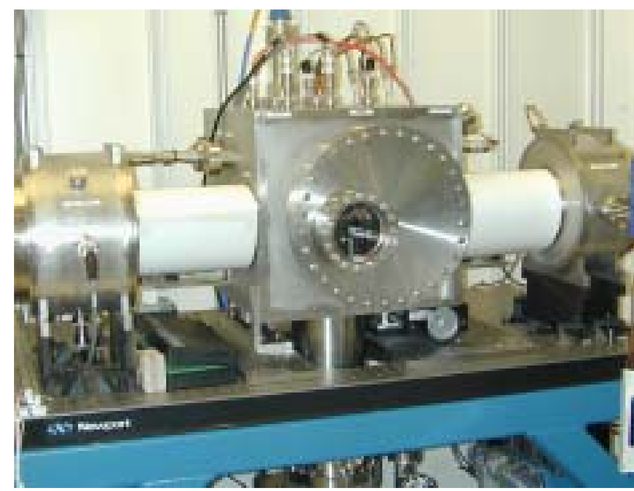
4 reflections deviatoric strain tensor

+ 1 energy full strain tensor



3-D Polychromatic Microscope has 5 key Elements

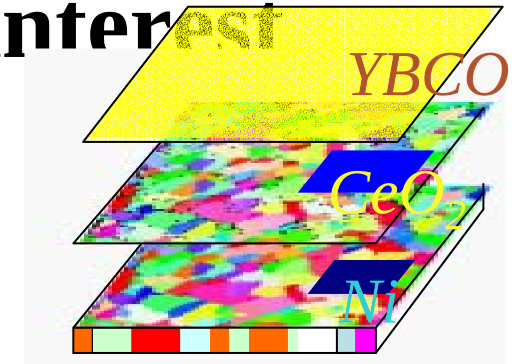




Operational 3D X-ray crystal m

- 10-22 keV
- Differential aperture microscopy with software

Programmatic mission centered on three areas of long-standing interest

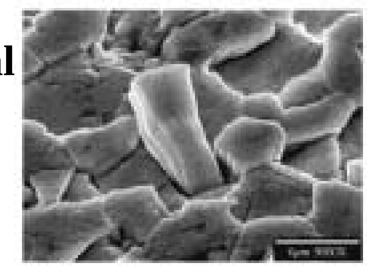


- Grain-growth
  - Epitaxial (near surface)
  - True 3D

- Deformation and strain localization
  - Mesoscale deformation using nanoindents
  - In-situ deformation in polycrystals

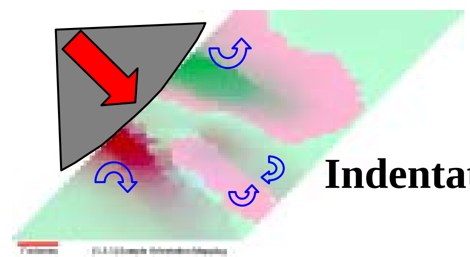
uickTime™ and a Video decompressor are needed to see this picture.

Al Polycrystal



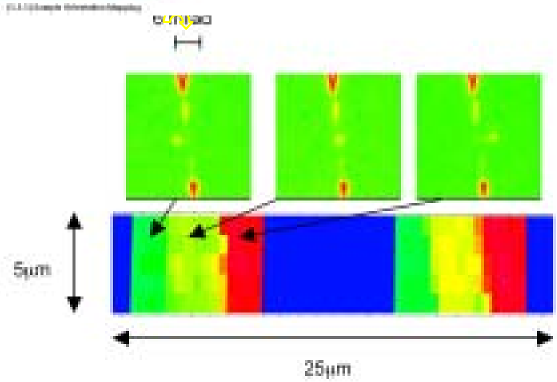
Tin Whiskers

- Fatigue and fracture
  - Thin films
  - Artificial cracks



Indentation

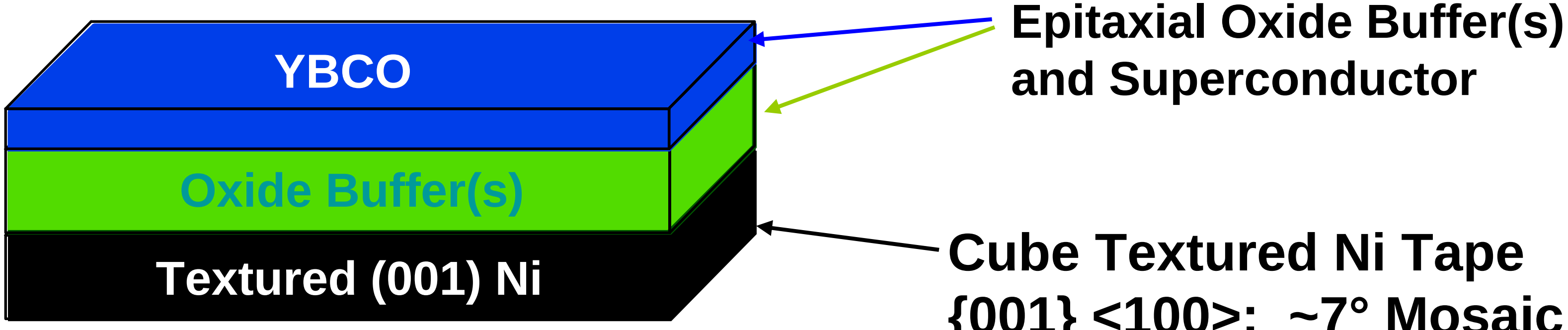
Compelling applications to many materials



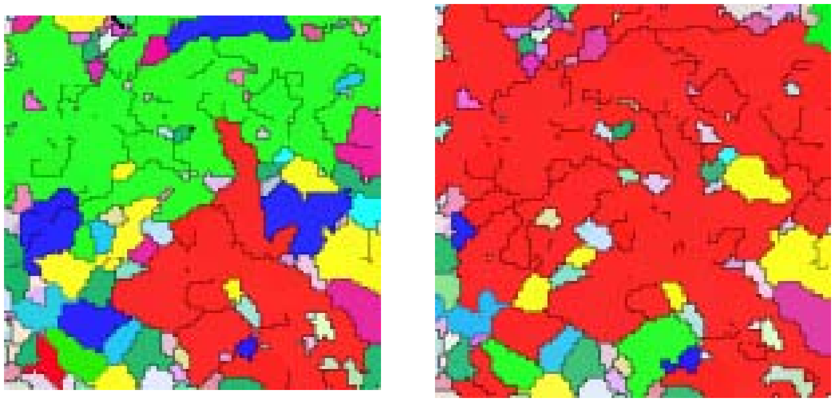
GaN on SiC

Rolling-Assisted Biaxially-Textured Substrates (RABiTS) practical approach High T_c Wires

RABiTS Architecture



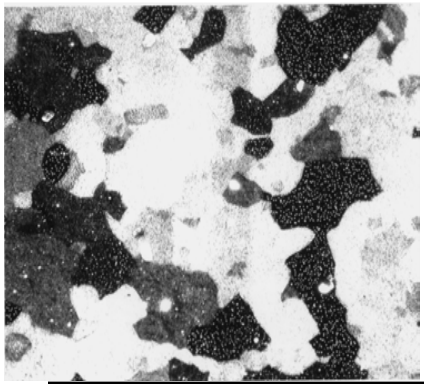
- Texture controls current transport
- Texture can be improved by buffer
- Scale-up requires fundamental understanding of epitaxial growth.

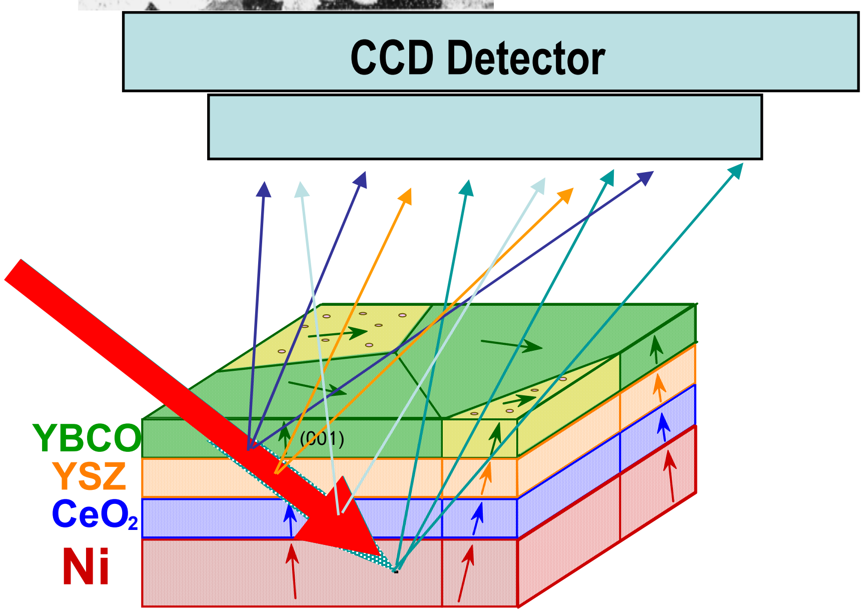


Ni CeO_2

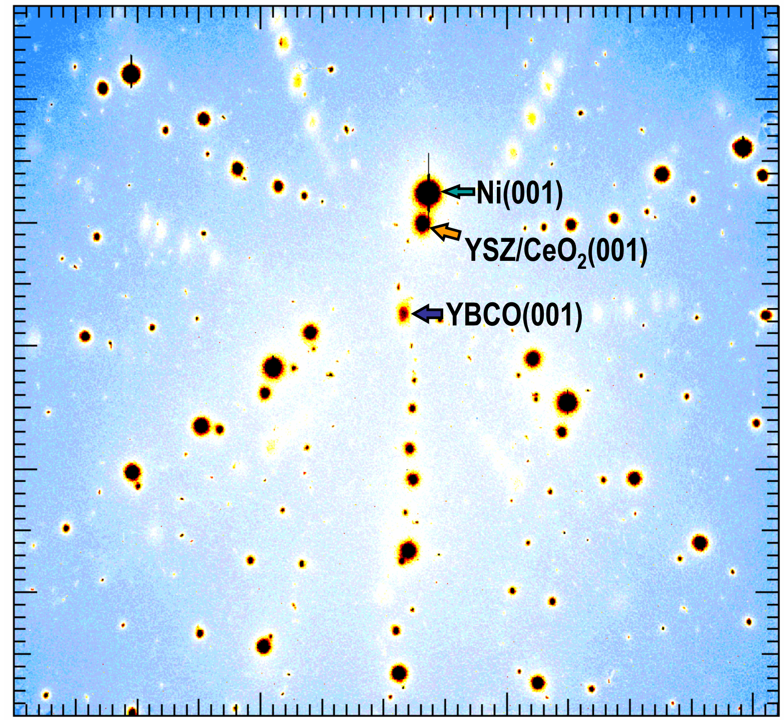
Budai et al. polychromatic microdiffraction to epitaxial growth RABiTS

Optical: ~50μm grains

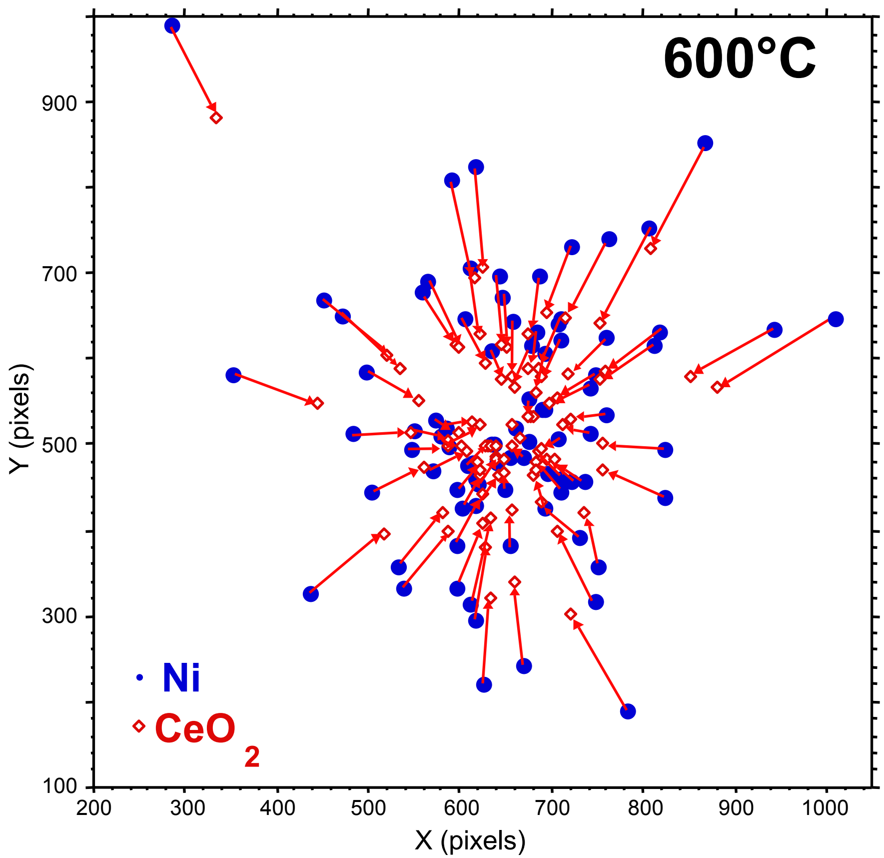


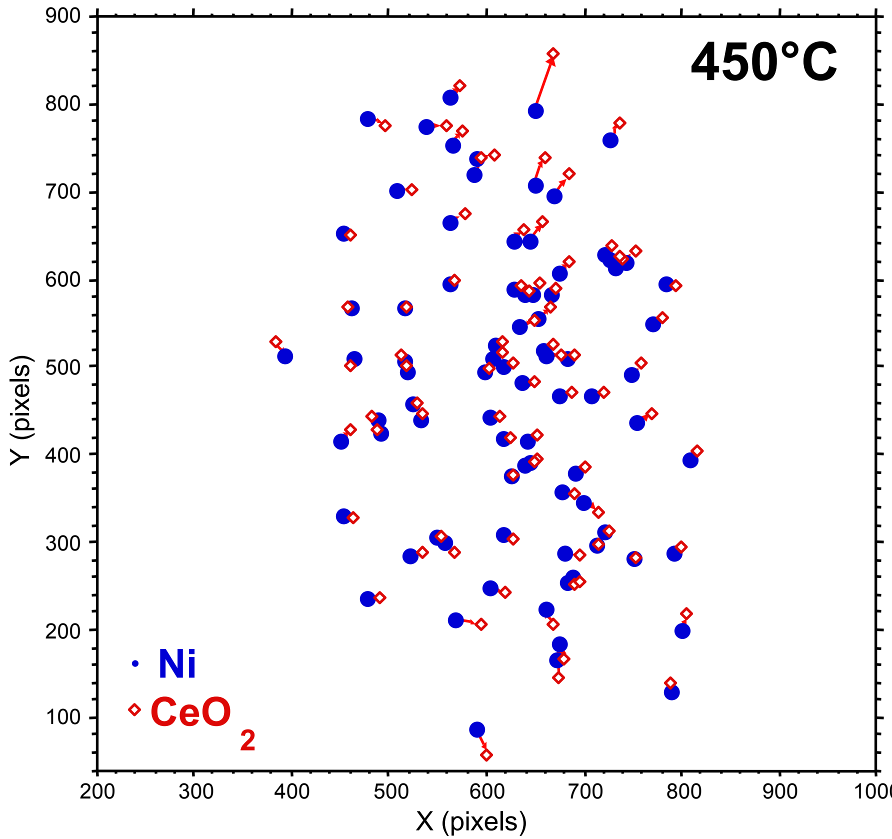


CCD Laue Patterns



Relative CeO_2 orientation depends deposition temperature





High temperature growth:

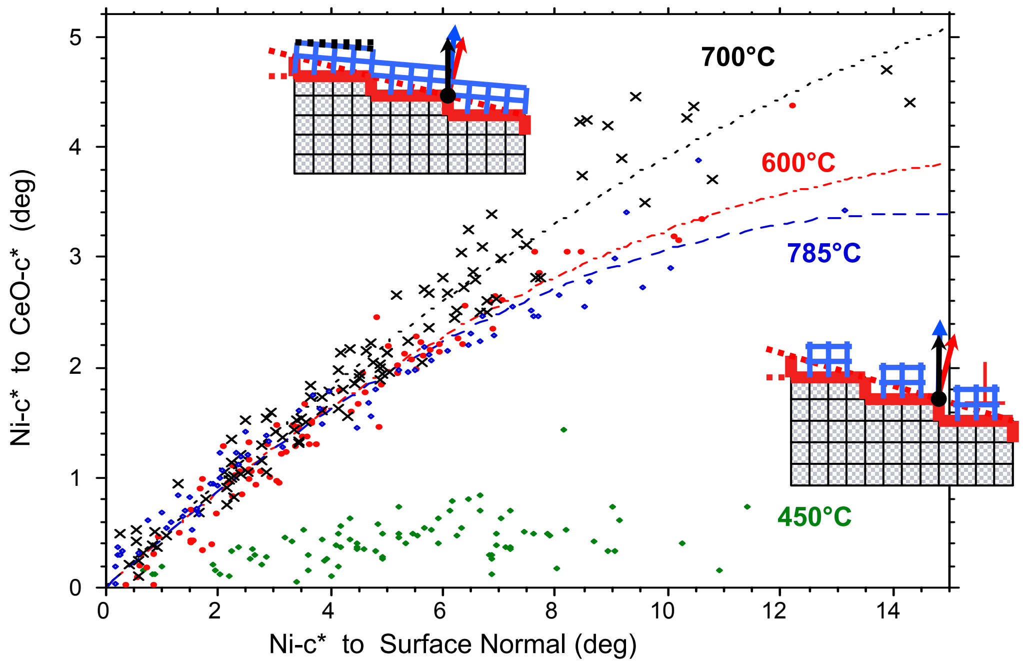
Crystallographic tilt towards ⊥

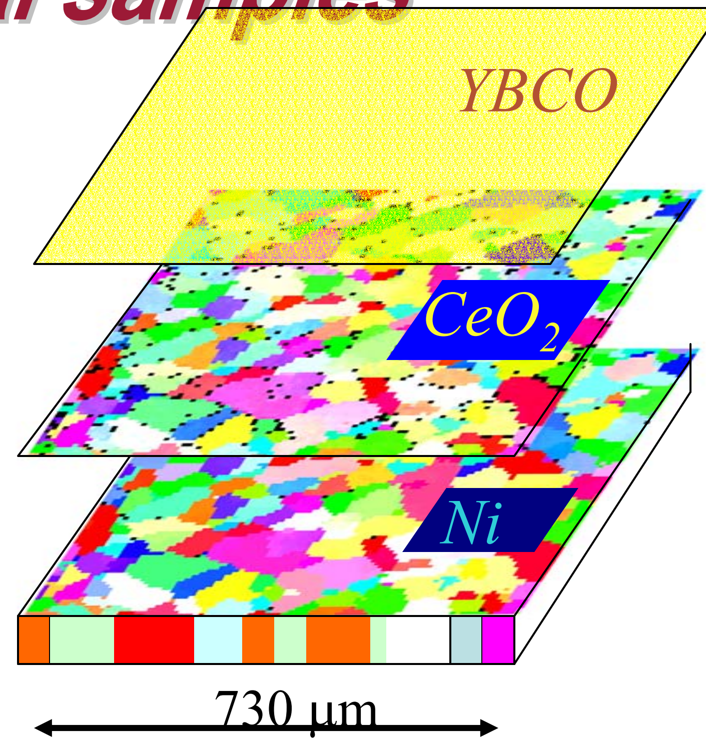
Tilt increases monotonically with miscut

Low temperature growth:

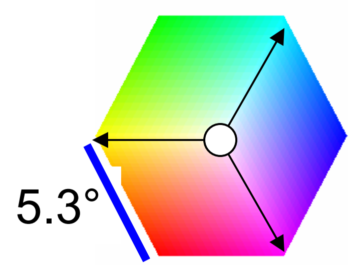
Small, ~biased tilts

Microbeam enables combinatorial measurements on real samples



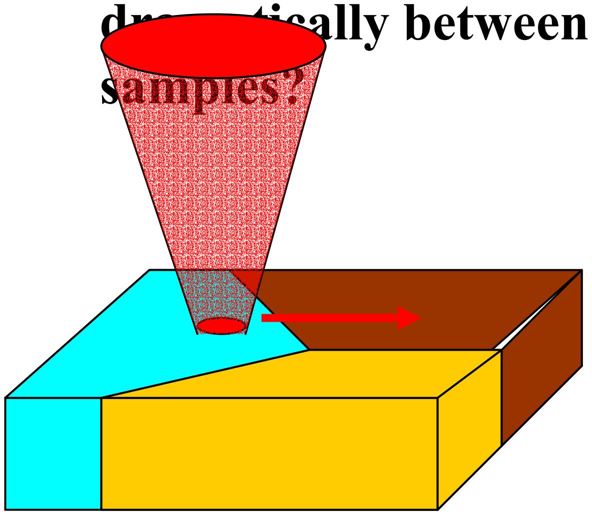


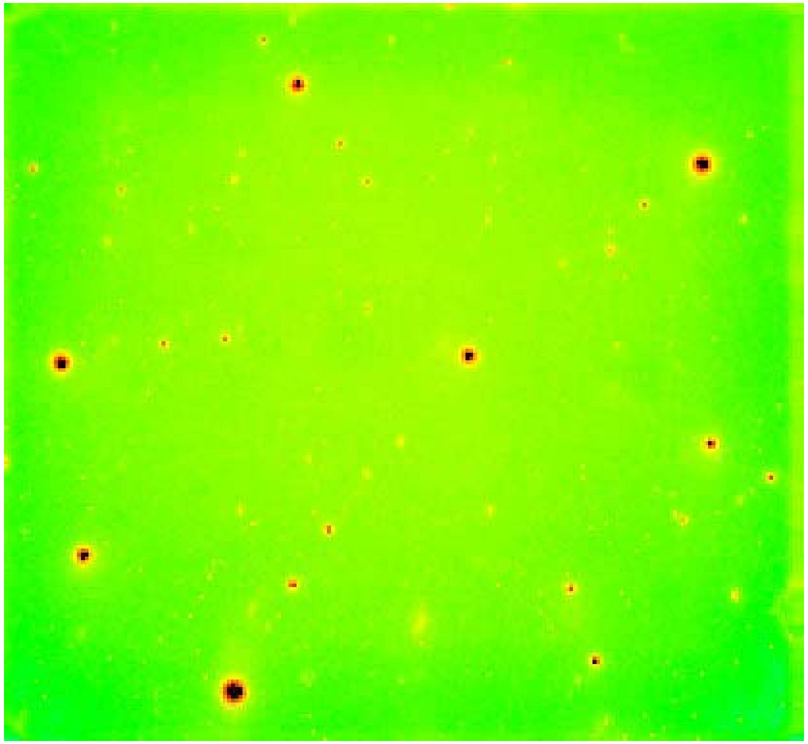
Budai JD, Yang WG, Tamura N, Chung JS, Tischler JZ, Larson BC, Ice GE, Park C, Norton DP **NATURE MATERIALS** 2 (7): 487-492 JUL 2003



Important questions remain

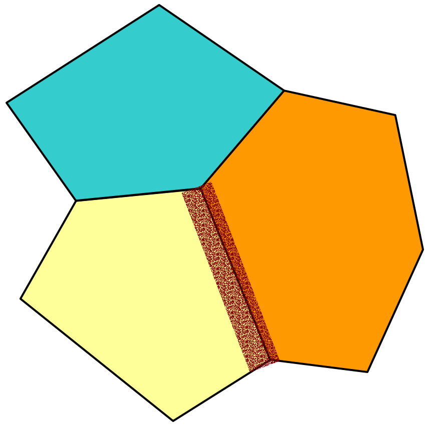
- **Why does J_c decrease for thick samples?**
- **Why does mosaic on single Ni substrate grain differ dramatically between samples?**

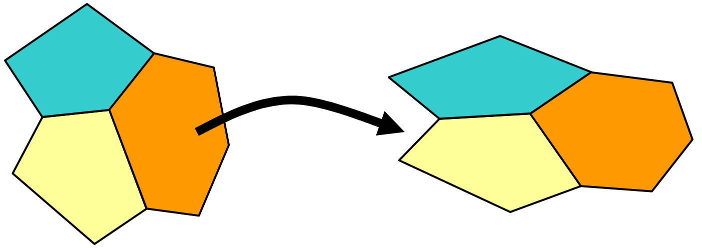




How grain boundary/polycrystal networks interact - a major materials opportunity 21st century

- What are the constitutive equations at grain boundaries?
  - How do they change with boundary type
- What are ideal microstructures?
  - How do different networks evolve during processing and in service?
- How can grain boundary distributions be controlled?
  - Grain boundary engineering

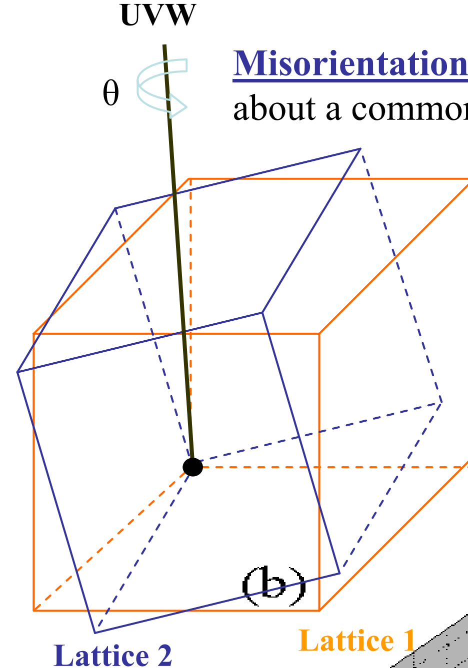




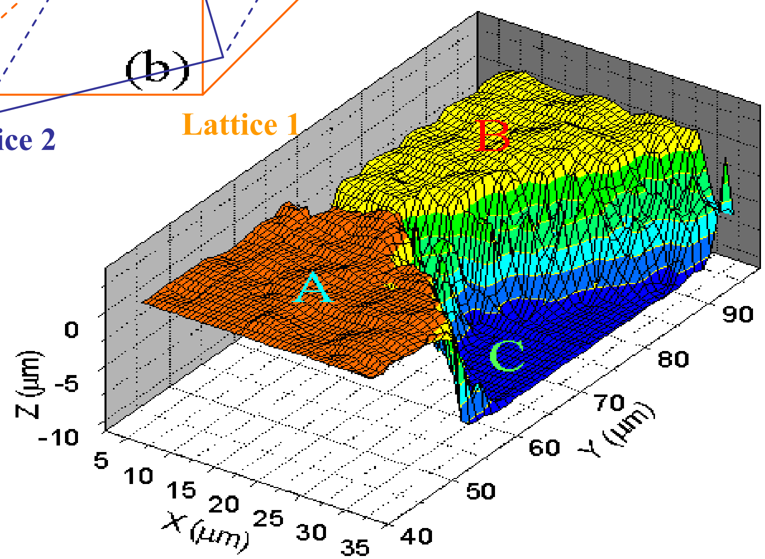
Essential for nanophase and advanced layered materials

Unprecedented precision addresses long-standing issues/ tests CSL models

- **CSL low-energy boundaries share lattice sites**
  - **Σ denotes inverse fraction of shared sites**
  - **Theory: misorientation increases as Σ *decreases***
- **Measured misorientation increase *with* Σ**
- **Grain boundary normals**
  - **Ideal directions should have lower energy**
  - **Faceting may remove energy advantage**

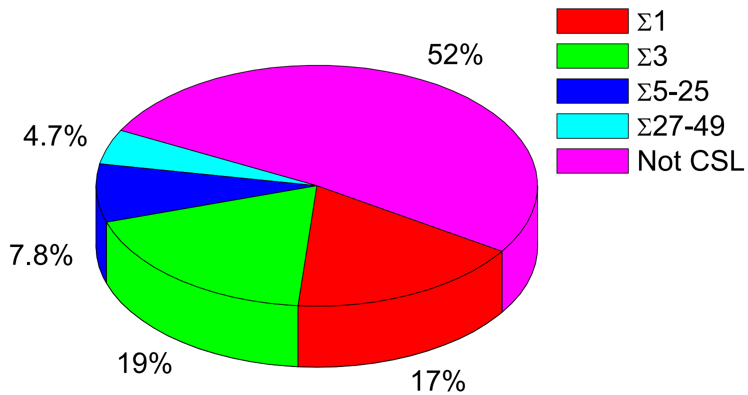


Misorientation: rotation of θ angle about a common axis UVW

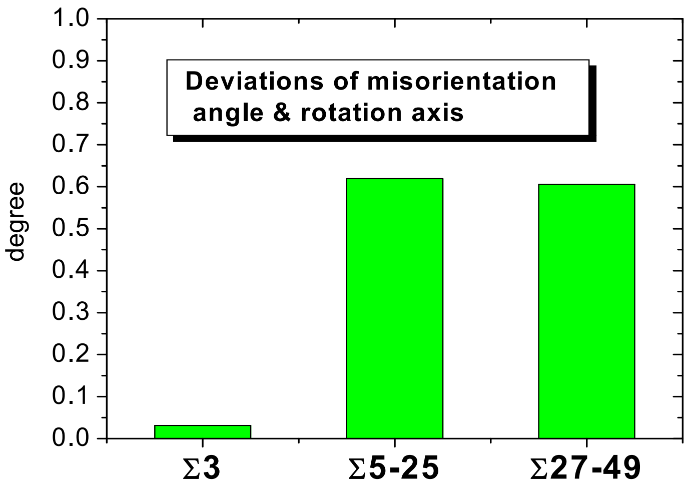


Morphology of Ni triple junction

Significant statistical information emerging



Total: 70



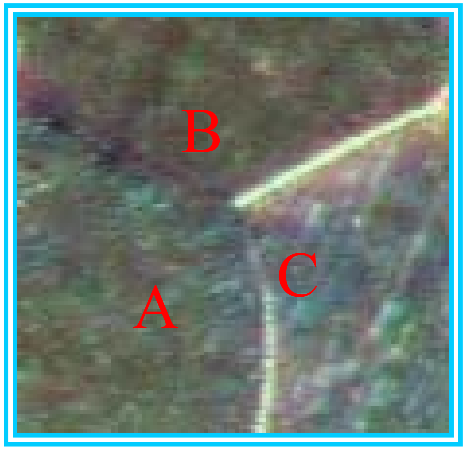
About 50% are CSLs, and 20% are found to be tilt, twist or having low-index in both grains.

No	Sigma type	Rotation Angle (degree)	Rotation angle off (degree)	Rotation Axis (RAX)	Rotation axis off (degree)	Boundary Normal (BN) in bi-crystal	Angle between RAX – BN (degree)	
B2	Σ21b	44.40	0.01	2, 1, 1	2.95	1.00, 0.32, 0.30 / 0.69, 1.00, 0.17	86.3	Tilt
B6	Σ47b	43.66	0.80	3, 2, 0	6.11	1.00, 0.07, 0.53 /1.00, 0.87, 0.31	74.6	Tilt
B10	Σ37c	50.57	0.14	1, 1, 1	4.55	0.08, 1.00, 0.26 /1.00, 0.12, 0.68	57.6	
B34	Σ1	(6.16°)				0.00, 1.00, 0.17 / 0.04, 0.27, 1.00	88.3	Tilt
A57	Σ3	60.00	0.01	1, 1, 1	0.02	1.00, 0.11, 0.02 / 0.32, 1.00, 0.87	86.4	Tilt
A314	Σ3	60.00	0.01	1, 1, 1	0.03	0.28, 0.31, 1.00 /0.36, 0.37, 1.00	2.4	Twist

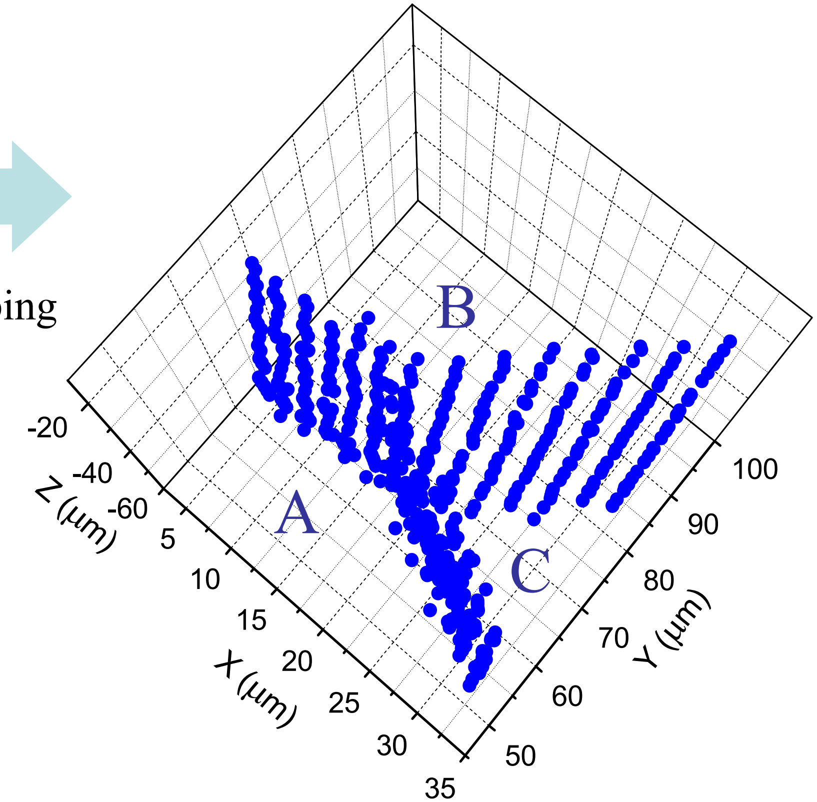
Open questions:

1. *Why and how are the deviations from ideal CSL model as Σ type increases?*
2. *Are there residual strains imposed near the deviated CSL boundaries?*
3. *Any difference of CSLs between near or below sample surface?*
4. *… … … …*

Three Dimensional Morphology of Triple Junction

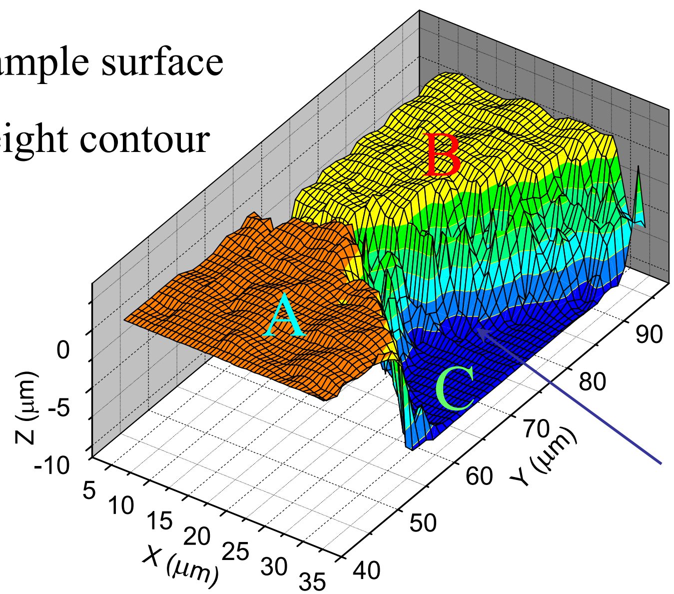


3-D mapping



Sample surface

height contour



surface step between two grains

Misorientaion angles:

A-B: 16.572°

B-C: 12.907°

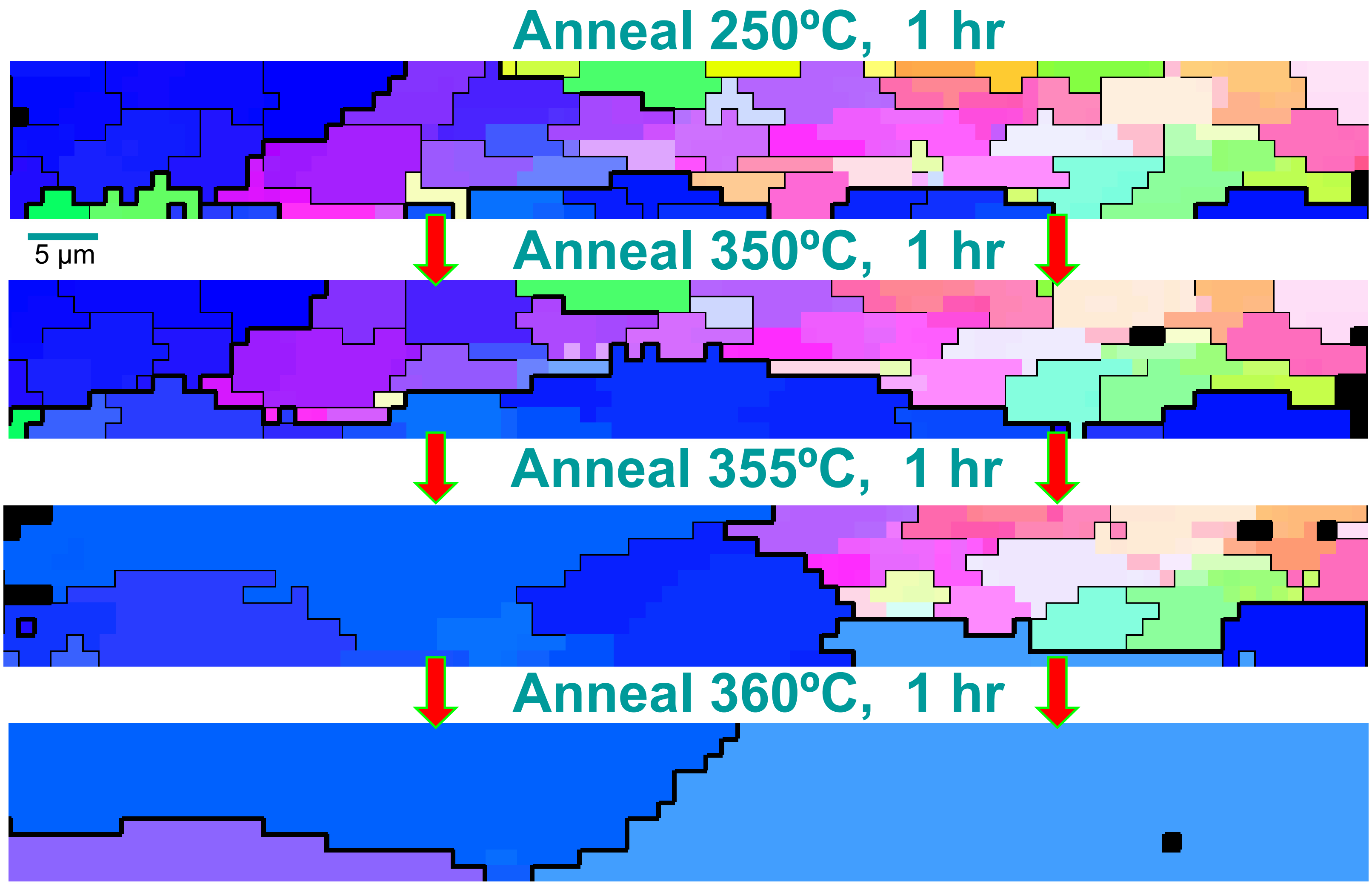
C-A: 5.538°

Polycrystalline grain structure can now be measured nondestructively in 3D-submicron resolution-meso scale

QuickTime™ and a
Video decompressor
are needed to see this picture.

Thermal Grain Growth in Hot-Rolled Aluminum

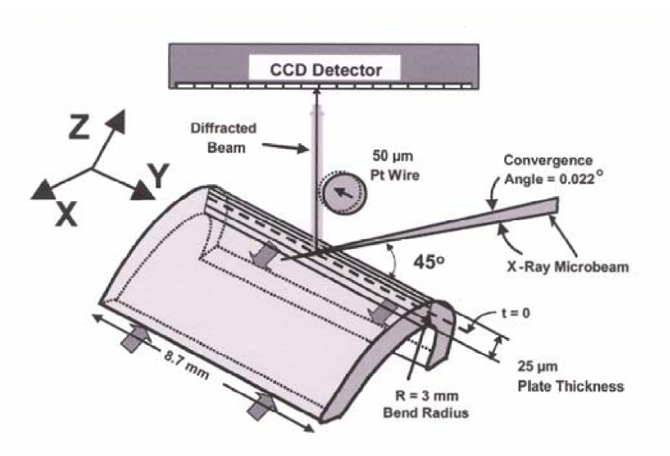
1 µm pixels,   Boundaries: 5° & 20°

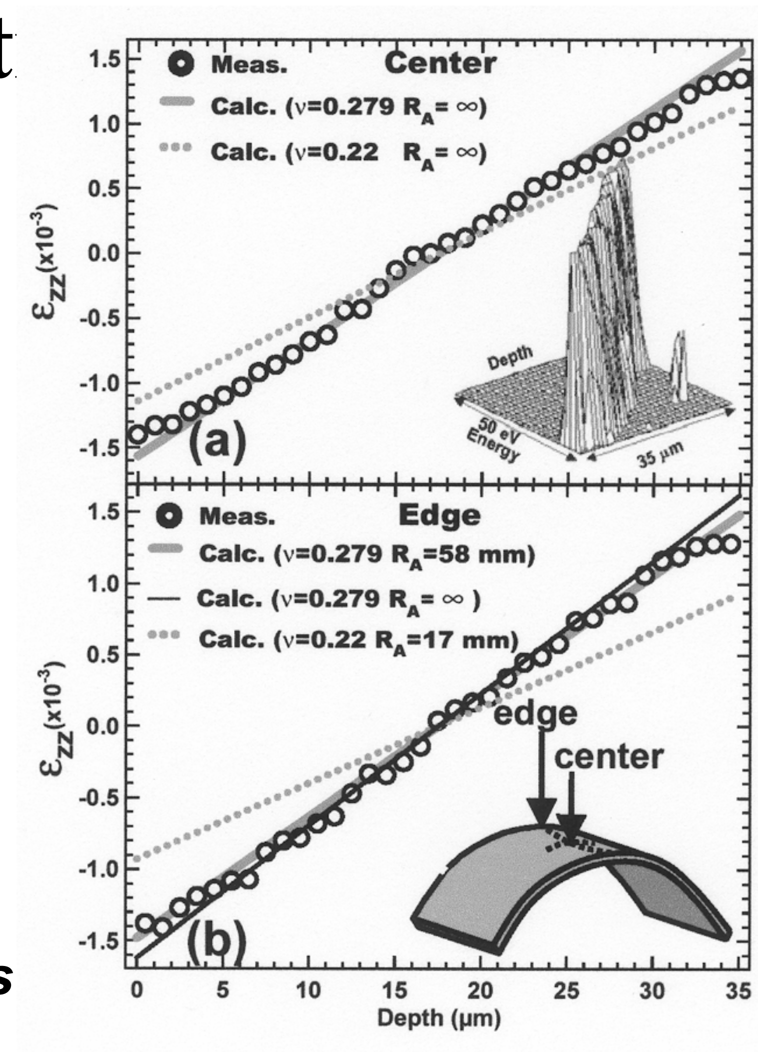


- GB motions include both high-angle and low-angle boundaries
- Complete and detailed 3D evolution needed for validation of theories.

Elastic strain key driving force-Monochromatic DAXM measures intra-granular elast

- Local strain-even in single crystal
- Ultra-high precision local orientations
- Independent of grain orientation
- Phase sensitive

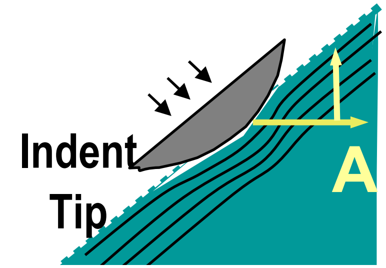




Revolutionizes ability to study materials

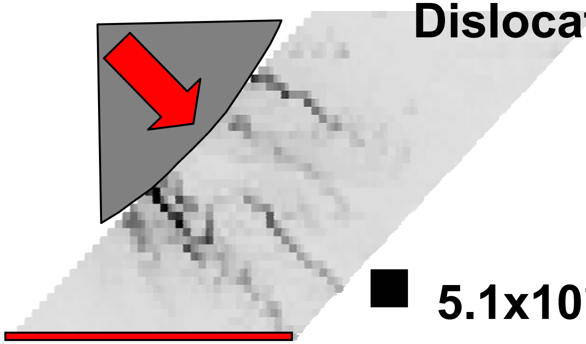
Nanoindent in single crystals provides major insights into 3D deformation/modeling

- Deformation boundary conditions completely known/ volume modelable
- Best models predict some features not others-highly reproducible
- Single, bi-crystal, or polycrystal
- Strain-gradient models directly testable



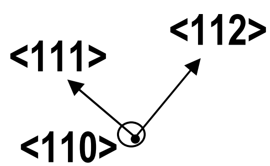
Probe Geometry

Dislocation Density Map

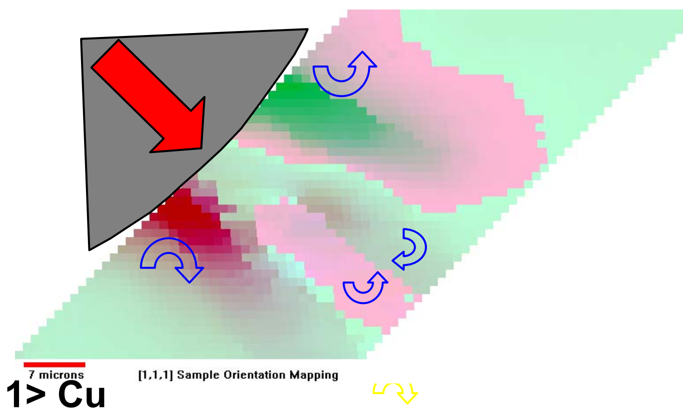


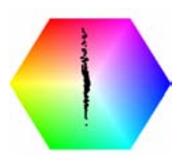
■ $5.1 \times 10^{10}/cm^2$

40 μm



Lattice Rotation Map





5 °

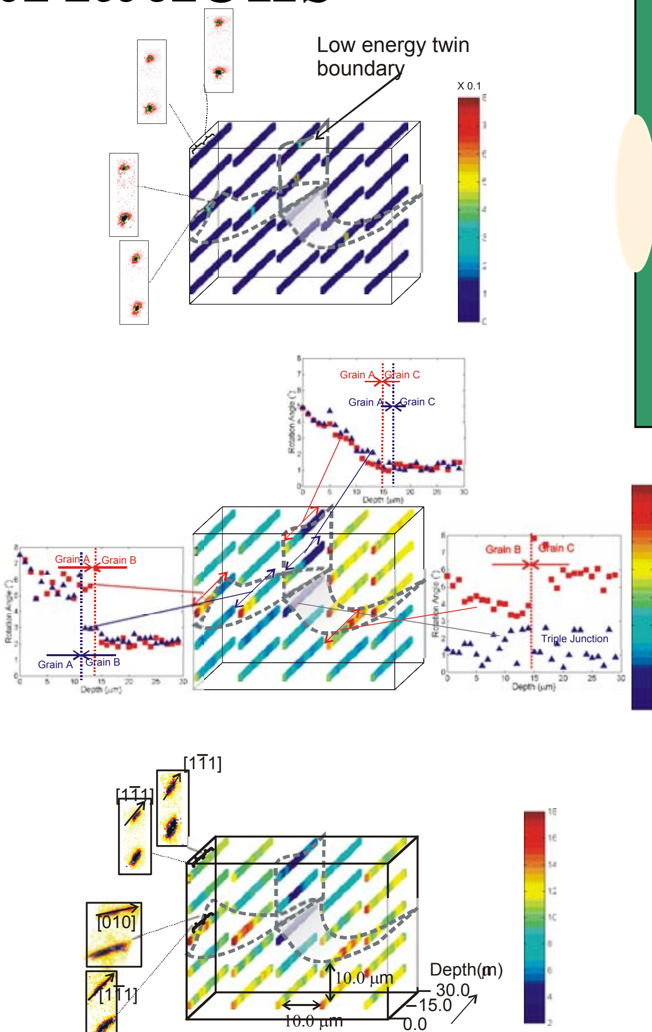
<111> Cu
100 mN Indent Force
69 μm Indent Radius

In-situ tensile deformation polycrystal finds intra-granular variations

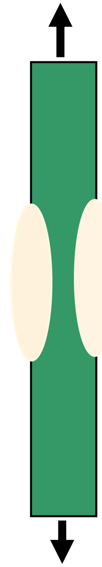
- Dramatic changes in deformations within single grain
  - Consistently large rotations near surface
- Plastic *and* elastic deformation measured
  - Essential information for understanding mechanisms
- Extensive sample characterization required for full boundary conditions

Proposed research

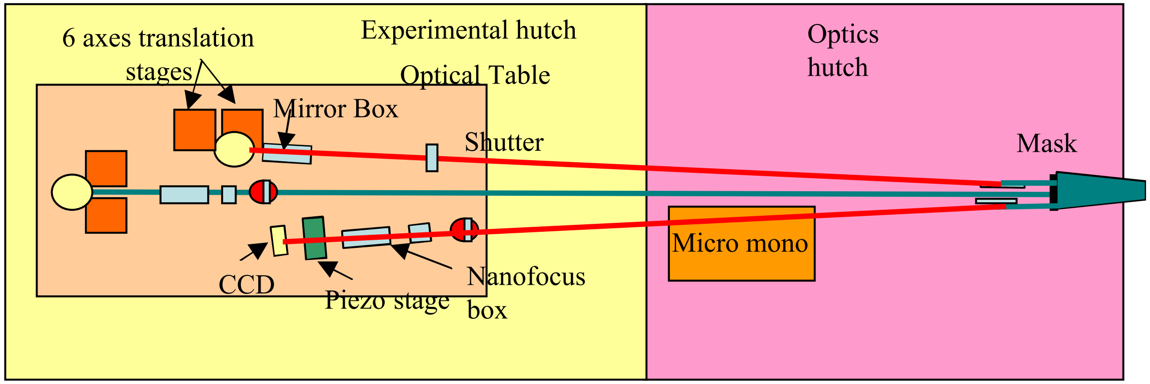
- **Full boundary conditions**
- **Low deformation**
- **Integrate theory**



Tensile deformed poly-Ni



To achieve potential and meet emerging demand - new microbeam lines and hardware proposed



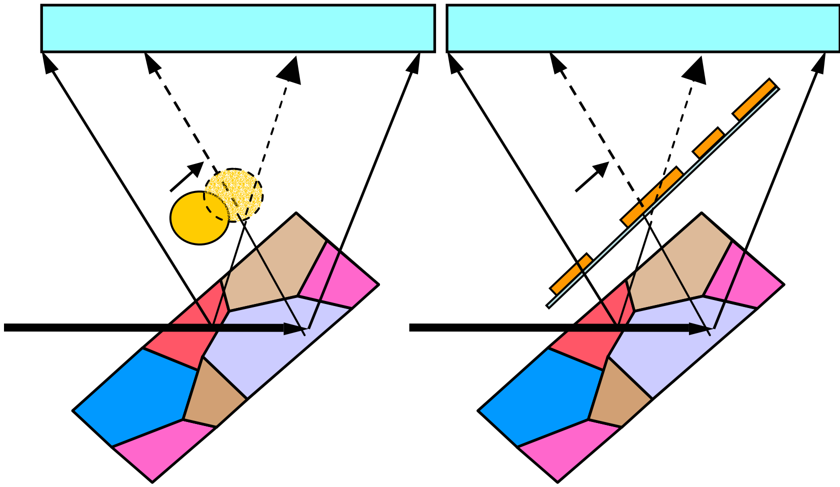
Multiplexed 3D polychromatic diffraction-center for mesoscale research-

- **-BM**
- **-Operated by APS**
- **-Greater general user access**

Spatial resolution 50nm→10nm

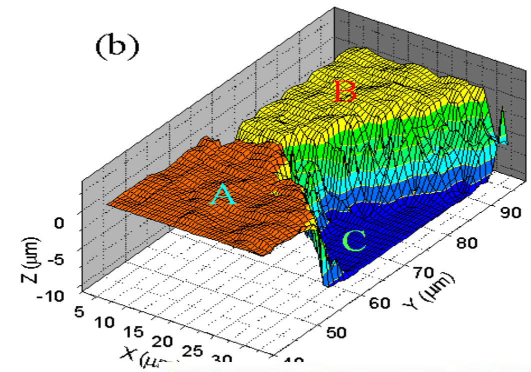
Accelerated 3D characterization 100-1000x

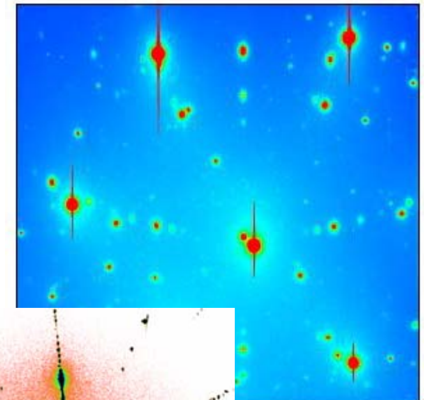
- **- Multiple wire/coded aperture**
- **- Faster detectors (GE detectors)**

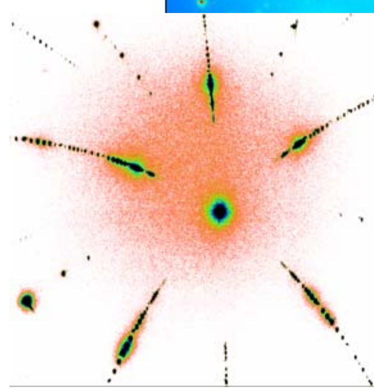


Summary: - important emerging technique

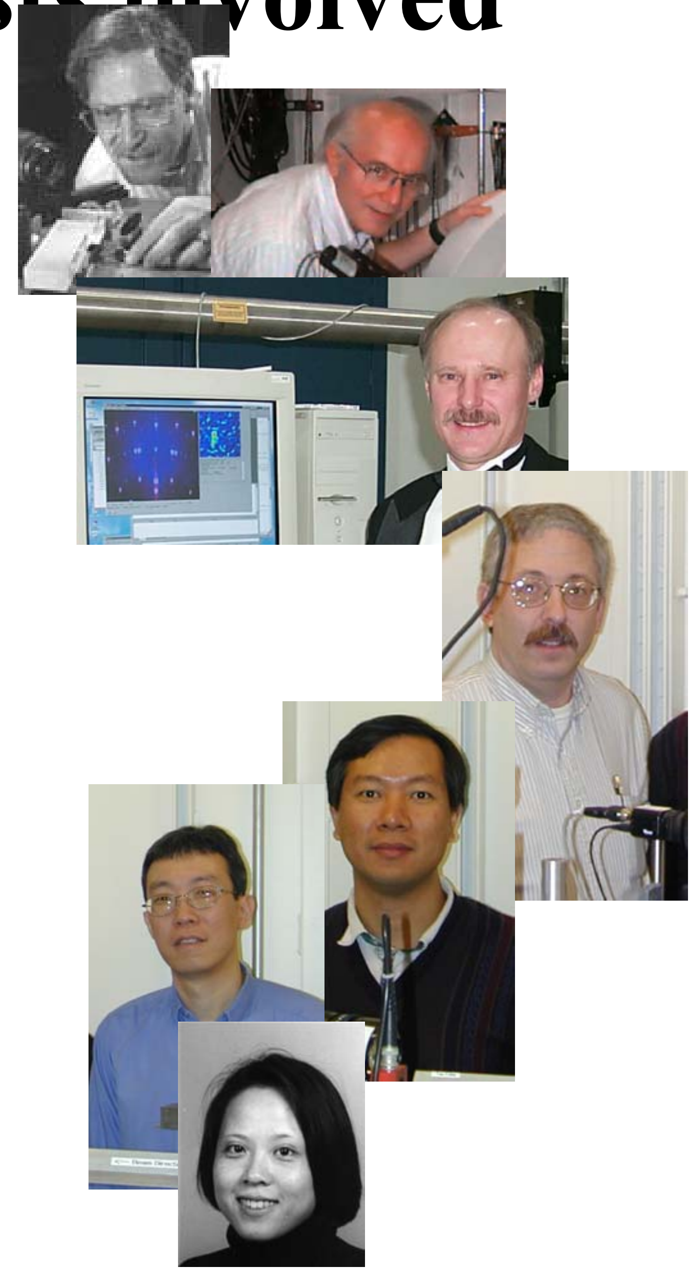
- Cannot meet demand with existing facilities
- Addresses long-standing issues with fundamentally new approach
- Wide applicability







Team of ORNL scientists involved



- Gene Ice- **Co-principle investigator, x-ray optics**
- Bennett Larson- **Co-principle investigator-3D deformation/nanoindentation**
- John Budai-**Epitaxial films and 3D grain growth**
- Jonathan Tischler-**Mesoscale measurements and computer analysis (CMSD - APS Site)**
- Wenge Yang-**Mesoscale deformation using nanoindentation (Guest Scientist- APS Site)**
- Wenjun Liu-**Grain boundary networks (Post Doc-APS Site)**
- Judy Pang**-in-situ 3D polycrystalline deformation**

Important support from APS-differentially deposited elliptical mirrors and beam stabilization